Swiss halocarbon emissions for 2019 to 2020 assessed from regional atmospheric observations

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Introduction

Swiss halocarbon emissions for 2019 to 2020 assessed from regional atmospheric observations by Rust et al.

The paper presents the results of an observation/modelling activity aimed at estimating emissions, at the national scale, of a wide range of halocarbons which are ozone-depleting and/or radiatively active gases.

This activity is recognized as relevant from a policy perspective because useful for the validation of national emission inventories and to ascertain the countries' compliance to the international agreements.

The paper is well written and clear, and the research is based on an outstanding and well-established observation activity. However, I have reservations about the methodology used for data analysis and the interpretation of results.

RC1: Concerning the methodology, my first question is about the station network. It is well known that reliable regional modelling requires a dense network of stations, which in most regions is not available. As stated in the introduction, in Europe, the AGAGE network continuously measures halocarbons. Beside Jungfraujoch and Mace Head, the network includes two additional sites (Mt Cimone and Ny-Alesund). Moreover, Taunus station in Germany started its monitoring activity in the period covered by the present study. In total, within the study domain, there are three more sites measuring halocarbons in addition to those used by the authors for their analysis.
Considering that the Swiss territory is located between two complex and strong pollutants' source regions, Germany to the north and Italy to the south, and considering that both Mace Head and Tacolneston are far away from sources that might affect the signal at JFJ and BRM and are not able to trace transport from the south, it would be important if the authors could justify the choice of using a less dense network.
Reply: The decision of the selection of sites was, on the one hand, based on the quality and completeness of the observations at each site and, on the other hand, on the sensitivity of the sites to Swiss emissions, the main focus of this study. The sites used in this study employ the same Medusa measurement technique for the compounds presented here, whereas two of the other mentioned sites (Taunus and Monte Cimone) rely on alternative measurement techniques that do not result in the same set of reliably observed compounds as presented here. Furthermore, the sites Monte Cimone and especially Ny Alesund are not very sensitive to Swiss emissions. In case of the

former, the Alps form a natural barrier that considerably lowers sensitivities although the site is not very distant from Switzerland itself. It is true that Monte Cimone is sensitive to emissions in northern Italy and that there may be some cross talk from changes in those emissions to emissions in Switzerland, but we considered the much larger sensitivity of the Beromünster observations to outweigh such crosstalk by far. The observations from Ny Alesund are generally not very sensitive to European emissions and the site is generally not used for regional emission estimates in Europe. Concerning the observations from Taunus Observatory, we agree with the reviewer that these are very valuable for the European perspective. In order to analyse their impact on Swiss emission estimates, we ran an additional inversion for HFO-1234yf including continuous observations from Taunus Observatory. Swiss national emissions in this run differed by less than 5 % from the base case. Since HFO-1234yf emissions showed a typical distribution with large emissions in the Benelux area, Germany and Northern Italy, we don't expect the impact of additional observations from Taunus Observatory to have a larger impact for other compounds either.

RC1: Have the authors performed comparative tests to determine the sensitivity of the receptor to the source using different sets of stations (including those mentioned above)?

Reply: Next to the inversion run including observations from an additional site (Taunus Observatory) we also performed a sensitivity run excluding the sites from the British Isles. For HFO-1234yf the result for Swiss national emissions only differed from the base run by 3 %. However, we saw that including MHD and TAC had a strong impact on a posteriori emissions from the Benelux area and Western Germany. To avoid any kind of cross talk from northwestern Europe to Switzerland it was deemed beneficial to include the two sites in the inversions for all compounds. Further addition of Taunus Observatory had less impact on emissions from northwestern Europe. Hence, for additional reasons given above, Taunus was not included. The results discussed here for HFO-1234yf may not be the representative for all compounds investigated, but performing the same set of sensitivity inversions for all compounds was beyond the scope of the current analysis. In future studies, the selection of observational data should be checked again for specific compounds. We have added the following discussion of observation selection to the manuscript:

"Sensitivity inversions for HFO-1234yf were performed in order to quantify the sensitivity of the a posteriori emissions in Switzerland to the selection of measurement sites. When adding additional observations from the Taunus Observatory in central Germany or when removing the observations from the British Isles, changes in total Swiss emissions were smaller than 5 %. "

RC1: Concerning the presentation of results (section 3.2), I do not see any result obtained using the Bayesian inversion at JFJ, neither a comparison between results obtained by Bayesian inversion at BRM and at JFJ. Since the comparison of the results obtained with the two methods (TRM and BI) at BRM highlights relevant differences for many of the compounds considered, with the TRM mostly over-estimating with respect to the BI, it would be useful to ascertain if the same deviations are observed comparing the two methods at JFJ.

Reply: The Bayesian inversion combines observations from all mentioned sites to derive spatially resolved emissions. This is in contrast to using information from individual sites in the TRM. We did not perform inversions based on JFJ observations only, given their limited sensitivity to Swiss emissions. However, we agree that the label "BI from BRM" was misleading and have removed it from the manuscript, i.e. we corrected this throughout the manuscript and the supplement.

RC1: Comparing the averaged BRM-TRM/BRM-BI fluxes with values obtained at JFJ by TRM only is less meaningful than comparing the average of two methods at BRM with the average of two methods at JFJ.
Therefore, I would like to ask why fluxes evaluated through the Bayesian Inversion at JFJ are not reported in this paper.
Reply: As explained above, Bayesian inversions for Swiss emissions were never calculated based on JFJ data only. JFJ observations are not sufficiently sensitive to Swiss emissions alone. This fact was one of the main reasons for the additional measurement campaign at BRM. However, additionally including JFJ (and MHD, TAC) observations in this study helps the inverse modeling to arrive at a more robust solution for Swiss emissions as well.

RC1: Concerning the interpretation of the results (page 17), the authors state that there is little consistency between BRM and JFJ data for a subset of three chlorinated gases. However, looking at percentage differences in Table 2, the lack of consistency seems to affect many compounds considered in the study.
Reply: On page 23 in the track changes document; Unfortunately this sentence on the lack of consistency towards the Jungfraujoch results was misleading. We meant to say that not only the Bayesian inversion and the Beromünster tracer-ratio results diverge from each other, but that also the Jungfraujoch tracer-ratio results diverge from the latter two. We re-wrote the paragraph to make it clear.

RC1: In the conclusions (page 22), the authors state that BRM data provide valuable information for the validation of halocarbon inventories. I agree that the use of a not remote station located in the middle of the Swiss territory at an altitude of 700 m, allows the authors to investigate emissions from the Swiss boundary layer with higher reliability.

However, given the large differences in fluxes evaluated using two different methods, the interpretation of results is quite difficult, and there is a risk that the analysis, rather than supporting the importance of observation-based methods, might lead to some doubts for most of the compounds considered in the study, except for the three most emitted HFCs (134a, 125 and 32) or for the sum of the HFCs.
Reply: On page 28 in the track changes document; Thank you for this justified remark. This issue was also extensively addressed in the reply to the second independent reviewer. To meet this point, we adjusted the last sentence of the conclusions and added another subsection (Section 3.2.4 Methods appraisal) to the manuscript discussing the two calculation methods in this light: "Both applied methods, the TRM and the BI, have their advantages and disadvantages. For the TRM we make the assumptions that the analyte and the tracer have similar spatial and temporal emissions sources, and that the transport distance is either sufficiently short for the ratio of analyte and tracer to be preserved until reaching the receptor, or that the transport distance is sufficiently long so that analyte and tracer emissions from multiple sources are well-mixed when reaching the receptor (Sect. 2.4). The Bayesian inversion makes the assumption that emissions are constant in time. For compounds with intermittent emissions, this may lead to reduced model performance. Furthermore, the method seeks to locate emissions in space, guided by a priori information. In the case of large spatial differences between a priori and real emissions, the method will be challenged, once again leading to reduced model performance. We observe, that for most substances, except the major HFCs, the HFOs, and SF_6, the TRM result exceeds the BI result. Possible reasons for this are that the assumption of similar emission sources of analyte and CO as the tracer does not hold and/or that the analyte and tracer emissions are not well mixed when reaching the receptor, leading to a distortion of the halocarbon-tracer

ratio. Nonetheless, we used CO as a good universal tracer for many substances. If we used another dispersedly emitted tracer, we would have similar problems. For both calculation methods, we indicated the reliability of the emissions results (Sect. 2.4, 2.5, and Fig. 4) and regarding the most highly emitted substances, we especially consider our results dependable for the major HFCs, the three HFOs, and SF_6."

Specific minor comments

RC1: Pag 8, line 256: the reference Reimann et al., 2020 to be checked or added

New line numbering "track_changes" document: P.9, L.297

New line numbering "corrected" document: P.8, L.285

Reply: We updated it to the publication of Reimann et al. (2021).

RC1: Pag 10, line 344: I would use “distributed” rather than calculated

New line numbering "track_changes" document: P.12, L.421

New line numbering "corrected" document: P.11, L.379

Reply: done

RC1: Pag 13, line 416: please consider adding the reference Keller et al. (2011) “Evidence for under-reported western European emissions of the potent greenhouse gas HFC-23”

New line numbering "track_changes" document: P.17, L.507

New line numbering "corrected" document: P.14, L.461

Reply: done. We added the Keller et al. (2011) reference.

Additional corrections by the authors, apart from the reviewer comments:

Abstract: We improved the abstract text so that it reads better

Jungfraujoch-based emission estimates: We updated the reference of (Reimann et al. 2020) to (Reimann et al. 2021) and updated the Jungfraujoch-based emission values to this report, or, where needed, to the newest calculation results for the corresponding years, as these emission values are adjusted and improved constantly for the year before the newest report is published. This is because the Jungfraujoch-based emissions are calculated as a three-year average. The difference to the values listed in this manuscript before adjustment are small, however.

Beromünster tracer-ratio method: We used the underlying data-set of carbon monoxide (CO) acquired by the Swiss NABEL network. The instrument is another version of Picarro analyzer, also using cavity ring-down spectroscopy. For the tracer-ratio emission results this makes only a minor difference, but we changed the method description for CO measurements in Section 2.2 "Sampling and Analysis" and updated the new emission results in Table 2.

Bayesian inversion: We added more details on the source of the a priori values for specific substances in the text and in the caption of Table 1. The a priori values listed in Table 1 were updated to the originally used UNFCCC and CLIMGAS values, not the a priori values already modified by the inversion calculations, since this might be confusing. The emissions results in Table 2 were updated in the context of the changed modelling.

Acknowledgements: We added more details regarding the contributors and funding of this study.

A few minor corrections: We corrected single words or punctuation characters throughout the manuscript.